Observations of Melting in Nanometer-Sized "Crucibles"

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Combining the use of the unique electron beam microcharacterization facilities at Berkeley and Argonne, a team of scientists has made the first direct high-resolution observations of melting and solidification in bimetallic alloys. By confining nanometer-sized "ingots" in a solid matrix, the researchers were able to characterize the initial stages of liquid formation on a distance scale two orders of magnitude smaller than previously feasible.

Background - Although the melting and solidification of solids are macroscopic phenomena, the early stages of both processes involve poorly understood interactions at the near-atomic scale. For this reason, a full understanding of these important phenomena has been difficult to achieve. Most observations of melting are made on a micrometer scale by optical or scanning electron microscopy which are limited to surfaces. By comparison, TEM is able to observe melting on a nanometer scale and at internal interfaces. This gives researchers access to true phase equilibria that are impossible to obtain otherwise.

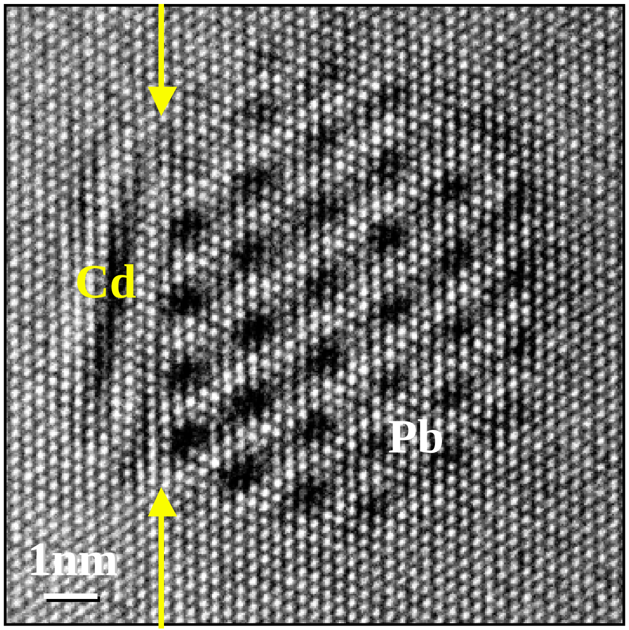
Accomplishment – Using the specialized electron beam facilities at Berkeley and Argonne Labs, the team was able to make direct in-situ observations of the melting transition, first by heating at constant composition and then by alloying at constant temperature. The results show that melting is initiated at special sites in the interface and display equilibrium contact angles characteristic of interfacial energies.

"Ingots" of a low-melting lead-cadmium alloy embedded as inclusions in a high-melting aluminum matrix (the "crucible") were made by special processing techniques. A high resolution image of such an alloy inclusion shows that at room temperature the ingots have distinct segments of pure lead and pure cadmium, each with their own, characteristic crystal structures.

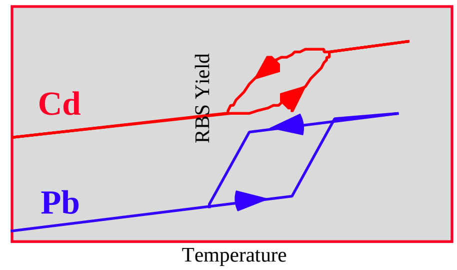
Bulk analysis by RBS/channeling showed distinctly different melting and solidification behavior for Pb and Cd. Direct observation during in-situ heating showed the lead-cadmium alloy to initiate melting at the triple line where the three metals are in contact (see figure and schematic). Examination of many such melting events also allowed the team to measure the characteristic angle of contact between the liquid Cd-Pb alloy and the solid Al matrix, which is a measure of the interfacial energy.

The group performed complementary in-situ alloying experiments using Argonnne National Laboratory's Intermediate Voltage Electron Microscope. Pure lead inclusions in Al were held at a temperature below their melting point and observed in-situ in the microscope. The unique capability of the IVEM-Tandem Facility to provide simultaneous TEM observation and ion implantation was used to implant Cd ions while observing the structure of the inclusions. This work showed for the first time how melting of single-phase particles initiates at specific crystallographic facets as the Cd concentration in the inclusions is increased.

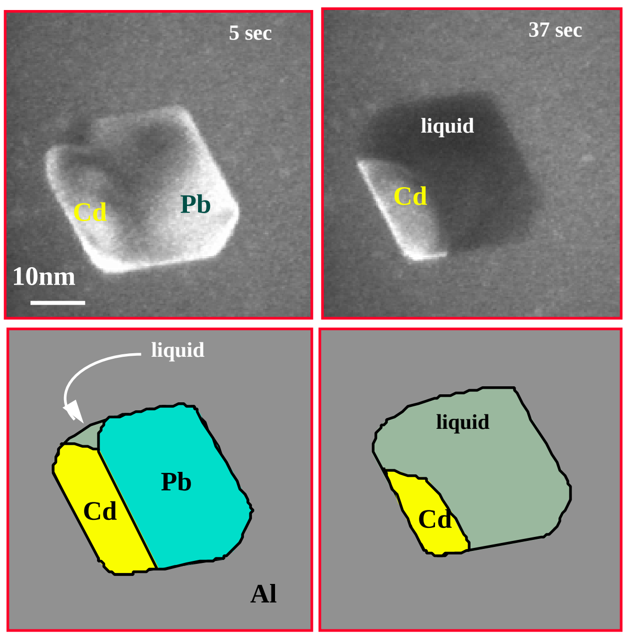
These observations will be of great value in the continuing study of fundamental questions concerning the mechanism of phase transformations. Alloy melting and solidification are also of importance in technologically important processes such as soldering in the semiconductor industry, welding in the steel industry and directional solidification in aerospace alloys.

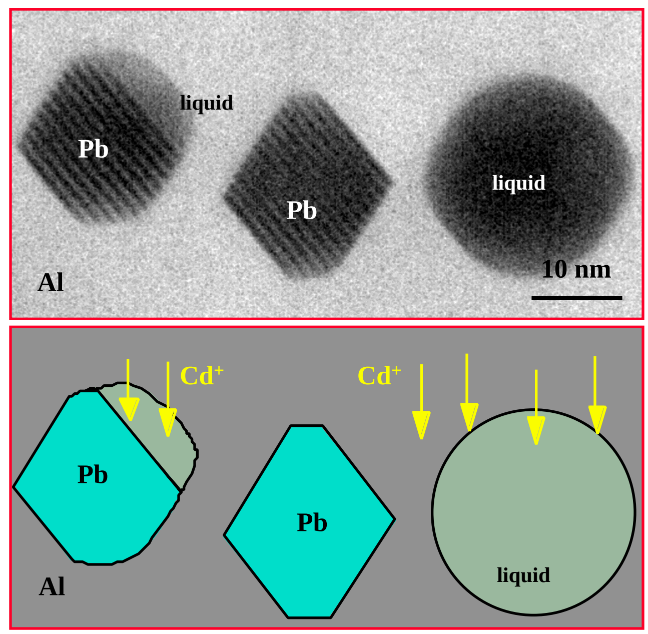


Left: Atomic resolution electron micrograph of nanoscale Pb-Cd “ingot” in Al matrix. The alloy separates into pure Pb and pure Cd regions, each with its own characteristic crystal structure. Arrows indicate internal interface between Cd and Pb.



Schematic of RBS/channeling analysis during heating/cooling cycle showing the difference in Pb and Cd segments of inclusions during melting and solidification





In-situ **heating** at constant composition

NCEM - LBNL

Melting observed in real time in the in-situ heating stage of NCEM’s analytical electron microscope. The melt nucleates at the triple junction (arrow) between Pb, Cd, and the Al matrix. Local contact angles are characteristic of interfacial energies.

In-situ **alloying** at constant temperature

IVEM Tandem Facility – ANL

Simultaneous Cd implantation and TEM observation allowed observation of melting as a function of inclusion composition at fixed temperature. Melting initiates at specific crystallographic facets as the Cd concentration in the inclusions is increased.

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